CNF-770645-1

To be published in Proceedings of the Fourth Rochester
Conference on Coherence and Quantum Optics
Rochester, New York - June 8-10, 1977

ADIABATIC FOLLOWING IN TWO-PHOTON TRANSITION

Munir H. Nayfeh and Ali H. Nayfeh

By acceptance of this article, the publisher or
recipient acknowledges the U.S. Government's right
to retain a non-exclusive, royalty-free license in
and to any copyright covering the article.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

1144

Adiabatic Following in Two-Photon Transition

Munir H. Nayfeh*

Oak Ridge National Laboratory**
Oak Ridge, Tennessee 37830

and

Ali H. Nayfeh

Virginia Polytechnic Institute and State University
Blacksburg, Virginia 24061

More interest has been generated recently in coherent multiphoton transitions in many-level systems¹⁻⁵. Coherent two-photon propagations, exhibiting self-induced transparency when twice the propagating frequency is resonant with a two-level system, have been analyzed¹⁻², and the one- and two-photon resonant fields were shown to obey together an area theorem². Tan-no et al³ reported the observation of coherent propagations based on a two-photon transition which involved the interaction of two optical pulses of different frequencies with a gaseous three-level system. Further, self transparency in a resonant two-photon absorption with a two-level system was observed⁴. In this work, we consider the effect of relaxation on the response of a three-level system to a smoothly varying near resonant, two-photon field.

Consider a three-level system (Fig. 1) with only level 2 optically connected to the ground state and the resonance frequencies ω_{12} and ω_{23} are far away from each other. The interaction of two smooth pulses with the above system can be described by the equation of motion of the density matrix

*Permanent address: Department of Physics, Yale University, New Haven, Connecticut 06520 (effective August 15, 1977).

**Operated by the Union Carbide Corporation for the Energy Research and Development Administration.

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [H, \rho], \quad (1)$$

where $H = H_0 + H'$, H_0 is the unperturbed Hamiltonian, and H' is the interaction Hamiltonian which can be written as

$$H_1 = \begin{bmatrix} 0 & v_{12} & 0 \\ v_{21} & 0 & v_{23} \\ 0 & v_{32} & 0 \end{bmatrix}, \quad (2)$$

where $v_{12} = \mu_{12} \cdot E_1$, $v_{23} = \mu_{23} \cdot E_2$, and the electric field is given by

$$\underline{E} = \underline{E}_1 + \underline{E}_2 = \underline{E}_1 \cos \omega_1 t + \underline{E}_2 \cos \omega_2 t. \quad (3)$$

Note that E_1 and E_2 were taken to induce transitions between the levels 1-2 and 2-3, respectively, due to the assumption that ω_{12} is much different from ω_{23} . The equation of motion of the diagonal and off-diagonal elements of the density matrix are derived by substituting Eq. (2) in Eq. (1). The result is

$$\dot{\rho}_{33} = -\frac{i}{\hbar} (v_{32}\rho_{23} - \rho_{23}^* v_{23}), \quad (4)$$

$$\dot{\rho}_{11} = -\frac{i}{\hbar} (v_{12}\rho_{12}^* - \rho_{12} v_{21}), \quad (5)$$

$$\dot{\rho}_{13} = -i(\omega_{12} + \omega_{23})\rho_{13} - \frac{i}{\hbar}(v_{12}\rho_{23} - \rho_{12}v_{23}), \quad (6)$$

$$\dot{\rho}_{23} = -i\omega_{23}\rho_{23} - \frac{i}{\hbar}(\rho_{33} - \rho_{22})v_{23} - \frac{i}{\hbar}v_{21}\rho_{13}, \quad (7)$$

$$\dot{\rho}_{12} = -i\omega_{12}\rho_{12} - \frac{i}{\hbar}(\rho_{22} - \rho_{11})v_{12} - \frac{i}{\hbar}v_{32}\rho_{13}. \quad (8)$$

The conservation law for the number of atoms implies that

$$\rho_{11} + \rho_{22} + \rho_{33} = 1. \quad (9)$$

Neither of the fields is taken to be resonant with the corresponding transitions so that level 2 is sufficiently far away from resonance, and thus its amplitude can be taken to be slowly varying. We write

$$\rho_{23} = \sigma_{23}e^{-i\omega_{23}t}, \quad \rho_{12} = \sigma_{12}e^{-i\omega_{12}t} \text{ and } \rho_{13} = \sigma_{13}e^{-i(\omega_{12}+\omega_{23})t}, \quad (10)$$

integrate Eqs. (6) and (7) by parts, and in the rotating wave approximation obtain

$$\sigma_{23} = \frac{(\rho_{33} - \rho_{22})\epsilon_2\mu_{23}}{\hbar\Delta\omega_2} e^{i\Delta\omega_2 t} + \frac{\mu_{21}\epsilon_1\sigma_{13}}{\hbar\Delta\omega_1} e^{i\Delta\omega_1 t} \quad (11)$$

$$\sigma_{12} = \frac{(\rho_{22} - \rho_{11})\epsilon_1\mu_{12}}{\hbar\Delta\omega_1} e^{i\Delta\omega_1 t} + \frac{\mu_{32}\epsilon_2\sigma_{13}}{\hbar\Delta\omega_2} e^{i\Delta\omega_2 t}, \quad (12)$$

where $\Delta\omega_1 = \omega_1 - \omega_{12}$ and $\Delta\omega_2 = \omega_2 - \omega_{23}$. The transformation of σ_{23} and σ_{12} in terms of σ_{13} results in an induced two-photon polarization which, when substituted in Eqs. (4) - (6) results in an explicit form for the two-photon coupling. In evaluating the two-photon coupling and the induced level shifts, we evaluate the amplitudes of σ_{23} and σ_{12} when $|\Delta\omega_2| = |\Delta\omega_1| = \Delta\omega_0$, which implies a slowly varying condition of the off-resonant frequency $\Delta\omega_0$ with respect to the virtual state 2. Substituting Eqs. (11) and (12) in (4) and (5) and using the rotating wave approximation, we obtain

$$\frac{d}{dt} (\rho_{33} - \rho_{11}) = k\epsilon^2 [i(\sigma_{13} - \sigma_{13}^*)\cos\Delta_0 t - (\sigma_{13} + \sigma_{13}^*)\sin\Delta_0 t], \quad (13)$$

$$\frac{d}{dt} (\sigma_{13} + \sigma_{13}^*) = k\epsilon^2 (\rho_{33} - \rho_{11})\sin\Delta_0 t + \frac{(\mu_{12}^2\epsilon_1^2 + \mu_{23}^2\epsilon_2^2)}{4\hbar^2\Delta\omega_0} (\sigma_{13} - \sigma_{13}^*), \quad (14)$$

$$\frac{d}{dt} (\sigma_{13} - \sigma_{13}^*) = ik\epsilon^2 (\rho_{33} - \rho_{11})\cos\Delta_0 t + \frac{i(\mu_{12}^2\epsilon_1^2 + \mu_{23}^2\epsilon_2^2)}{4\hbar^2\Delta\omega_0} (\sigma_{13} + \sigma_{13}^*), \quad (15)$$

where $\Delta_0 = \Delta\omega_1 + \Delta\omega_2$ is the two-photon resonance detuning, $\epsilon^2 = \epsilon_1\epsilon_2$,

and $k = \frac{\mu_{23}\mu_{12}}{2\hbar^2\Delta\omega_0}$.

Equations (13) - (15) can now be written in terms of the Bloch vector components by defining

$$\begin{aligned} w &= \rho_{33} - \rho_{11}, \quad \sigma_{13} + \sigma_{13}^* = u \cos(\Delta_0 t) + v \sin(\Delta_0 t) \\ \sigma_{13} - \sigma_{13}^* &= -i[(u \sin(\Delta_0 t) - v \cos(\Delta_0 t))], \end{aligned} \quad (16)$$

where w is the two-photon inversion and u and v are the dipole dispersive and absorptive components, respectively. Substituting Eq. (16) in Eqs. (13) - (15) and equating the coefficients of $\cos\Delta\omega$ and $\sin\Delta\omega$, we obtain

$$\frac{du}{dt} + \gamma_2 u = -\Delta(t)v, \quad (17)$$

$$\frac{dv}{dt} + \gamma_2 v = \Delta(t)u + k\epsilon^2 w, \quad (18)$$

$$\frac{dw}{dt} + \gamma_1 w = -k\epsilon^2 v - \gamma_1, \quad (19)$$

where $\Delta(t) = \Delta_0 + s_1\epsilon_1^2 + s_2\epsilon_2^2$; $s_1 = \frac{\mu_{12}^2}{4\hbar^2\Delta\omega_0}$; $s_2 = \frac{\mu_{23}^2}{4\hbar^2\Delta\omega_0}$; and $\gamma_1 = 1/T_1$ and $\gamma_2 = 1/T_2$ are the atomic energy and phase relaxation widths, respectively.

Damping effects have been incorporated in the above equations phenomenologically by introducing a longitudinal relaxation time T_1 of the upper level and a homogeneous transverse relaxation time T_2 . The relaxation effects of the intermediate level may be neglected if it is sufficiently far away from resonance.

Note that the interaction of two pulses with a three-level system, when the intermediate state is sufficiently far away from resonance, can be described by the above "two-photon Bloch equations". These are analogous to the equations governing a one-photon transition in a two-level system, except for the presence of a two-photon coupling and a frequency shift⁵.

An adiabatic following model based on the above approximation was used to derive and explain parametric generation processes⁶. Power-dependent nonlinear susceptibilities were obtained, and pulse propagation was studied. However, in the above study, relaxation of the atomic

levels was neglected. In this paper we determine the effect of relaxation on the model and calculate the relaxation-dependent contributions to the nonlinear refractive index (n). The leading contribution to $(n-1)\varepsilon^{-2}$ is intensity dependent caused by the level shifts inherent in multiphoton processes; it includes a relaxation-dependent part which is important at times shorter than γ_1^{-1} . The second-order contributions depend on the square of the intensity and the time-integrated square of the intensity. The latter contribution, which is relaxation dependent, causes line asymmetry at the long wavelength wing; it consists of a term that is proportional to $(\gamma_2 - \gamma_1)$ and only important at early times and a term that is proportional to $(2\gamma_2 - \gamma_1)$.

In the intervals in which the amplitudes of the fields vary slowly with time, approximate solutions of Eqs. (17) - (19) can be obtained by using either the method of multiple scales⁷ or the factorization technique⁸⁻¹⁰ in which one first reduces these equations to three uncoupled third-order equations with time-dependent coefficients. To use the latter technique, we first eliminate the variable v from Eq. (19) by multiplying it by $k\varepsilon^2(\frac{d}{dt} + \gamma_2) \frac{1}{k\varepsilon^2}$ and then substituting for $(\frac{d}{dt} + \gamma_2)v$ from Eq. (18). The result is

$$[(\frac{d}{dt} + \gamma_2 - \frac{d}{dt} \ln k\varepsilon^2)(\frac{d}{dt} + \gamma_1) + k^2\varepsilon^4]w = -k\varepsilon^2\Delta u - k\varepsilon^2(\frac{d}{dt} + \gamma_2)\frac{\gamma_1}{k\varepsilon^2}. \quad (20)$$

The variable u can be eliminated from Eq. (20) by multiplying it by $k\varepsilon^2\Delta(\frac{d}{dt} + \gamma_2) \frac{1}{k\varepsilon^2\Delta}$ and substituting for $(\frac{d}{dt} + \gamma_2)u$ from Eq. (17) and for v from Eq. (19). We neglect terms the order of $\frac{d^2}{dt^2} \ln \varepsilon$, $(\frac{d}{dt} \ln \varepsilon) \times (\frac{d}{dt} \ln \varepsilon^2\Delta)$, and $(\frac{d}{dt} \ln \varepsilon)^2$ in the homogeneous part of the equations and terms the order of $\frac{d}{dt} \ln \varepsilon$ in the sources. Terms the order of $\frac{d}{dt} \ln \varepsilon$ are kept in the homogeneous parts because their integrals over time

determine the solutions. Consequently, these terms are not negligible for nonconstant field amplitudes even as the rate of change of the amplitudes becomes arbitrarily small. Using a similar procedure for Eq. (17), one can show that the results for w and u are

$$\left\{ \frac{d^3}{dt^3} + [2Y_2 + Y_1 - \frac{d}{dt} \ln(k^2 \epsilon^4 \Lambda)] \frac{d^2}{dt^2} + [(Y_1 + Y_2)^2 - Y_1^2 + \Omega^2 - (Y_2 + Y_1) \frac{d}{dt} \ln(k^2 \epsilon^4 \Delta)] \frac{d}{dt} + Y_1(\Delta^2 + Y_2^2) - (k^2 \epsilon^4 + Y_1 Y_2) \frac{d}{dt} \ln(k \Delta \epsilon^2) + (Y_2 + \frac{d}{dt} \ln k^2 \epsilon^4) k^2 \epsilon^4 \right\} w = -Y_1(\Delta^2 + Y_2^2) \quad (21)$$

and

$$\left\{ \frac{d^3}{dt^3} + (2Y_2 + Y_1 - \frac{d}{dt} \ln k \epsilon^2 \Lambda^2) \frac{d^2}{dt^2} + [Y_1 + Y_2)^2 - Y_1^2 + \Omega^2 - (Y_1 + Y_2) \frac{d}{dt} \ln \Lambda - 2Y_2 \frac{d}{dt} \ln k \epsilon^2 \Delta] \frac{d}{dt} + Y_1(Y_2^2 + \Delta^2) + Y_2 k^2 \epsilon^4 + (Y_2^2 + \Delta^2) \frac{d}{dt} \ln k \epsilon^2 \Delta + (2\Delta^2 - Y_1 Y_2) \frac{d}{dt} \ln \Lambda \right\} u = Y_1 \Delta k \epsilon^2, \quad (22)$$

where

$$\Omega^2(t) = \Lambda^2(t) + k^2 \epsilon^4. \quad (23)$$

The general solution of Eq. (21) is the sum of a particular solution and a linear combination of three linearly independent homogeneous solutions, two of which are oscillatory. For adiabatically switched fields, the oscillatory solutions vanish and for small Y_1 and $Y_2 k^2 \epsilon^4 \Omega^{-2}$ the nonoscillatory solution is slowly varying. Thus, to determine an approximate solution for the case of adiabatically switched fields when Y_1 and $Y_2 k^2 \epsilon^4 \Omega^{-2}$ are small, we need to determine only the particular solution of Eq. (19) and its slowly varying homogeneous solution. This can be accomplished by dropping the second and third derivatives

of w , the products of γ_1 and the derivatives of w or ϵ , and the products of the derivatives of w and the derivatives of ϵ and Δ . Under these approximations, Eq. (19) becomes

$$(\gamma_2^2 + \Omega^2) \frac{dw}{dt} + [\gamma_1(\Delta^2 + \gamma_2^2) - k^2 \epsilon^4 \frac{d}{dt} \ln(k\Delta \epsilon^2) + (\gamma_2 + \frac{d}{dt} \ln k^2 \epsilon^4) k^2 \epsilon^4] w = -\gamma_1(\Delta^2 + \gamma_2^2) \quad (24)$$

Similarly, under similar approximations, Eq. (22) becomes

$$(\gamma_2^2 + \Omega^2) \frac{du}{dt} + [\gamma_1(\Delta^2 + \gamma_2^2) + \gamma_2 k^2 \epsilon^4 - (\Delta^2 + \gamma_2^2) \frac{d}{dt} \ln(k\epsilon \Delta^2) + 2\Delta \frac{d\Delta}{dt}] u = \gamma_1 \Delta k \epsilon^2 \quad (25)$$

We note that Eq. (24) can be derived alternatively by using the above approximations while eliminating u and v from Eqs. (17) - (19). Thus, this technique can be readily used to determine the influence of adiabatically switched fields on many level systems when the relaxations are small.

For an atom that is initially in the ground state, the solution of Eq. (24) is

$$w = - \frac{\Delta[\Delta_0^2 + \gamma_2^2]^{\frac{1}{2}}}{\Delta_0(\Omega^2 + \gamma_2^2)^{\frac{1}{2}}} e^{-\gamma_1 t - \Gamma(t)} - \frac{\gamma_1 \Delta}{(\Omega^2 + \gamma_2^2)^{\frac{1}{2}}} e^{-\gamma_1 t - \Gamma(t)} \int_{-\infty}^t \frac{\Delta^2 + \gamma_2^2}{\Delta(\Omega^2 + \gamma_2^2)^{\frac{1}{2}}} e^{\gamma_1 t + \Gamma(t)} dt \quad (26)$$

where

$$\Gamma = (\gamma_2 - \gamma_1) \int_{-\infty}^t \frac{k^2 \epsilon^4}{\Omega^2 + \gamma_2^2} dt + \gamma_2^2 \int_{-\infty}^t \frac{1}{\Delta(\Omega^2 + \gamma_2^2)} \frac{d\Delta}{dt} dt \quad (27)$$

When γ_2 is small, Eqs. (26) and (27) become

$$w = - \frac{\Delta(t)}{\Omega(t)} e^{[-\gamma_1 t - q(t)]} \gamma_1 e^{-q(t)} \frac{\Delta(t)}{\Omega(t)} \int_{-\infty}^t \frac{\Delta(\tau)}{\Omega(\tau)} e^{[\gamma_1(\tau - t) + q(\tau)]} d\tau, \quad (28)$$

where

$$q(t) = (\gamma_2 - \gamma_1) \int_{-\infty}^t \frac{k^2 \epsilon^4}{\Omega^2} dt. \quad (29)$$

When $\gamma_2 = 0(\Omega)$ and $k^2 \epsilon^4 \ll \Omega^2$, Eqs. (26) and (27) reduce to the rate equation limit

$$w = - e^{-\gamma_1 t - \tilde{\Gamma}(t)} \left[1 + \gamma_1 \int_{-\infty}^t e^{\gamma_1 t + \tilde{\Gamma}(t)} dt \right], \quad (30)$$

where

$$\tilde{\Gamma}(t) = \gamma_2 \int_{-\infty}^t \frac{k^2 \epsilon^4 dt}{\Delta_0^2 + \gamma_2^2}. \quad (31)$$

In this limit, the functions u and v are given in terms of w as

$$u = - \frac{\Delta_0 k \epsilon^2 w}{\Delta_0^2 + \gamma_2^2}, \quad (32)$$

$$v = \frac{\gamma^2 k \epsilon^2 w}{\Delta_0^2 + \gamma_2^2} \quad (33)$$

When the relaxations are not small, we cannot determine an approximate solution to Eq. (21) by using the above described simplified scheme. Instead, we use the factorization technique and express Eq. (21) as

$$\left(\frac{d}{dt} + g_3\right) \left(\frac{d}{dt} + g_2\right) \left(\frac{d}{dt} + g_1\right) w = - \gamma_1 (\Delta^2 + \gamma_2^2), \quad (34)$$

Equation (28) can be solved by letting $g_i = G_i + \delta_i$ where $\delta_i \ll G_i$.

Expanding Eq. (34) and comparing the zeroth- and first-order terms,

results in equations for the G_i and the amplitude functions δ_1 , δ_2 , and δ_3 . The equation for G_i is cubic. In some cases, this cubic equation has simple solutions. These include the case of equal decay widths, the case where the decay widths are smaller than or equal to $\nu(\frac{d}{dt} \ln \epsilon)$, and the case of small fields and small Y_1 . Note that, in the latter case, there is no restriction on the magnitude of Y_2 .

When $Y_1 = Y_2$, one finds¹⁰ that the general solution consists of two homogeneous solutions which oscillate with the Rabi frequency, a homogeneous, non-oscillatory solution, and a particular solution. For adiabatic fields and an atom that is initially in the ground state, the oscillatory solutions vanish and w takes the simple form

$$w = -\frac{\Lambda}{\Omega} e^{-\gamma t} - \gamma \frac{\Lambda}{\Omega} e^{-\gamma t} \left[\int_{-\infty}^t \frac{\Omega e^{\gamma \tau} (Y^2 + \Lambda^2)}{\Lambda(Y^2 + \Omega^2)} d\tau \right]. \quad (35)$$

When $Y_1 \neq Y_2$ it follows from Eq. (28) that the inversion has an induced damping $q(t)$. For weak fields, the lowest-order term in $q(t)$ depends on the integrated square of the intensity, an indication of the two-photon process. For sufficiently strong fields, $q(t)$ has no field dependence, and the inversion is damped only by the phase relaxation γ_2 , as in the one-photon transition. However, the population inversion given by Eq. (35) does not have any power-dependent decay. As $\gamma \rightarrow 0$, Eq. (35) reduces to

$$w \rightarrow -\frac{\Lambda}{\Omega}, \quad (36)$$

in agreement with the calculation of Ref. 6.

It is interesting to compare the above results with the results for two-level systems^{9,11,12}. The first contribution in Eq. (35) is identical

in form to the corresponding contribution in two-level systems, except that the detuning Δ is a function of time due to the frequency shift and the Rabi frequency Ω is modified by the two-photon coupling and the frequency shift. The presence of the frequency shift causes a shift in the crossing point of the inversion [$\Delta(t) = 0$]. However, the integral contribution form is different from that for two-level systems because Δ is a function of time in three-level systems.

Since Eq. (26) was derived with the assumption that $\gamma_2 k^2 \epsilon'' \ll \Omega^2$, which involves the product of γ_2 and $k^2 \epsilon''$, it is less restrictive on each of them individually. This implies that solution (26) is valid in the intermediate regime in which neither γ_2 nor $k^2 \epsilon''$ takes extreme values. Therefore, it bridges the gap between the rate equation limit (30) and the coherent limit.

In addition to the line shift, which is inherent in two-photon processes due to the involvement of virtual intermediate levels, there is a relaxation-dependent line asymmetry due to the buildup of the pulsed field. To see this, we derive an expression for the refractive index of three-level systems in the weak field limit. For an atom that is initially in the ground state, one can easily show that the solution of Eq. (25) is

$$u = \frac{k\epsilon^2}{\Omega} e^{-\gamma_1 t} - q(t) \left[1 + \gamma_1 \int_{-\infty}^t \frac{\Delta(\tau)}{\Omega(\tau)} e^{\gamma_1 \tau} + q(\tau) d\tau \right]. \quad (37)$$

for small γ_2 . When $\gamma_1 = \gamma_2 = 0$, u reduces to $k\epsilon^2/\Omega$, in agreement with the result of Ref. 6.

Since the refractive index n of the medium at either excitation frequency due to the nonlinear mixing of the field components is related to u by

$$n = 1 + 2\pi N k' u, \quad (38)$$

it is given for weak fields by

$$\begin{aligned} \frac{n-1}{\epsilon^2} = & - \frac{2\pi N k^2 \hbar}{\Lambda_0} \left[1 - \frac{s_1 \epsilon_1^2 + s_2 \epsilon_2^2}{\Lambda_0} + \frac{(s_1 \epsilon_1^2 + s_2 \epsilon_2^2)^2}{2\Lambda_0^2} - \frac{1}{2} \frac{k^2 \epsilon^4}{\Lambda_0} \right] \\ & \times \left(1 + e^{-\gamma_1 t} \right) - \frac{1}{2} (2\gamma_2 - \gamma_1) \int_{-\infty}^t \frac{k^2 \epsilon^4(\tau)}{\Lambda_0^2} e^{\gamma_1(t-\tau)} d\tau \\ & - (\gamma_2 - \gamma_1) e^{-\gamma_1 t} \int_{-\infty}^t \frac{k^2 \epsilon^4(\tau)}{\Lambda_0^2} d\tau + O(\epsilon^6). \end{aligned} \quad (39)$$

We note that the lowest-order nonlinear contribution to $(n-1)\epsilon^{-2}$ is intensity dependent. It is equal to $2\pi N k^2 \hbar \Lambda_0^{-2} (s_1 \epsilon_1^2 + s_2 \epsilon_2^2) [1 + \exp(-\gamma_1 t)]$ which is inversely proportional to the square of the frequency shift of the transition. The next-order contribution is $\pi N k^2 \hbar \Lambda_0^{-3} [k^2 \epsilon^4 - 2(s_1 \epsilon_1^2 + s_2 \epsilon_2^2) [1 + \exp(-\gamma_1 t)]]$; part of this contribution is due to the two-photon level shift. The last two contributions depend on the integrated square of the intensity. These contributions cause asymmetry in the refractive index at the long wavelength wing of the transition due to the accumulation of the square of the intensity at later times. We note that for pulses having the same total energy, the asymmetry depends on the pulse length. For long pulses, the term proportional to $(\gamma_2 - \gamma_1)$ vanishes, while the term proportional to $(2\gamma_2 - \gamma_1)$ increases with increasing times. Thus, it is expected that there exists a pulse length for which the asymmetry is maximum.

All of these contributions are dependent on the relaxation. In the absence of relaxation (i.e., $\gamma_1 = \gamma_2 = 0$), it follows from Eq. (39) that

$$\frac{n-1}{\epsilon^2} = - \frac{4\pi N k^2 \hbar}{\Lambda_0} \left[1 - \frac{s_1 \epsilon_1^2 + s_2 \epsilon_2^2}{\Lambda_0} + \frac{2(s_1 \epsilon_1^2 + s_2 \epsilon_2^2)^2}{2\Lambda_0^2} - \frac{k^2 \epsilon^4}{\Lambda_0} \right]. \quad (40)$$

When the relaxation is due entirely to spontaneous emission (i.e., $2\gamma_2 = \gamma_1$), the second contribution in Eq. (39) vanishes. In this case the two-photon

level shifts contribute to the asymmetry of the line to $O(\epsilon^4)$ at early times only. At times larger than the relaxation time T_1 , it follows from Eq. (39) that

$$\frac{n-1}{\epsilon^2} = -\frac{2\pi N k^2 \hbar}{\Delta_0} \left[1 - \frac{s_1 \epsilon_1^2 + s_2 \epsilon_2^2}{\Delta_0} + \frac{2(s_1 \epsilon_1^2 + s_2 \epsilon_2^2) - k^2 \epsilon^4}{2\Delta_0^2} \right. \\ \left. - \frac{1}{2} (2\gamma_2 - \gamma_1) \int_{-\infty}^t \frac{k^2 \epsilon^4(\tau)}{\Delta_0^2} e^{\gamma_1(\tau-t)} + q(\tau) d\tau \right]. \quad (41)$$

In conclusion, we have shown that the coherent interaction of two smoothly-varying, near-resonant, two-photon pulses with a three-level system can be described by "two-photon damped Bloch equations" which are analogous to those for a one-photon transition in a two-level system except for the presence of a two-photon coupling and a frequency shift. These equations are solved for the cases $\gamma_1, \gamma_2 \ll \Omega$, $\gamma_1 = \gamma_2$, $\frac{\gamma_2 k^2 \epsilon^4}{\Omega^2}$, and $\gamma_1 \ll \Omega$, where γ_1 and γ_2 are the atomic energy and phase relaxation widths and Ω is the Rabi frequency. The leading contribution to $(n-1)\epsilon^{-2}$ is intensity dependent caused by the level shifts inherent in multiphoton processes; it includes a relaxation dependent part which is important at times shorter than γ_1^{-1} . The second-order contributions depend on the square of the intensity and the time-integrated square of the intensity. The latter contribution, which is relaxation dependent, causes line asymmetry at the long wavelength wing; it consists of a term that is proportional to $(\gamma_2 - \gamma_1)$ and only important at early times and a term that is proportional to $(2\gamma_2 - \gamma_1)$. The case $\frac{\gamma_2 k^2 \epsilon^4}{\Omega^2}$, $\gamma_1 \ll \Omega$ bridges the gap between the coherent limit and the rate equation limit.

REFERENCES

- ¹E. M. Belenov and I. A. Polucktov, Sov. Phys. - JETP 29, 754 (1969).
- ²M. Takatsuji, Phys. Rev. A 4, 808 (1971).
- ³N. Tan-no, K. Yokoto, and H. Inaba, Phys. Rev. Lett. 29, 1211 (1972).
- ⁴T. L. Gvardzhadze, A. Z. Grasyuk, and V. A. Kovalenko, Sov. Phys. - JETP 37, 227 (1973).
- ⁵N. Tan-no, K. Yokoto, and H. Inaba, J. Phys. B 8, 339 (1975).
- ⁶D. Grischowsky, M. M. T. Loy, and P. F. Liao, Phys. Rev. A 12, 2514 (1975).
- ⁷A. H. Nayfeh, Perturbation Methods (Wiley-Interscience, New York, 1973), Chap. 6.
- ⁸M. G. Payne and M. H. Nayfeh, Phys. Rev. A 13, 595 (1976).
- ⁹M. H. Nayfeh, Phys. Rev. A 14, 1304 (1976).
- ¹⁰M. H. Nayfeh and A. H. Nayfeh, Phys. Rev. A 15, 1169 (1977).
- ¹¹R. H. Lehmberg and J. Reintjes, Phys. Rev. A 12, 2574 (1975).
- ¹²M. H. Nayfeh and A. H. Nayfeh, J. Appl. Phys. 47, 2528 (1976).

Figure Caption

Fig. 1. Energy level diagram of the three-level system.

ORNL-DWG 77-9405